

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
26 July 2001 (26.07.2001)

PCT

(10) International Publication Number  
**WO 01/54218 A2**

(51) International Patent Classification<sup>7</sup>: **H01M 8/02**,  
8/24, 8/04, B60L 11/18

(21) International Application Number: PCT/EP01/00438

(22) International Filing Date: 16 January 2001 (16.01.2001)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:  
60/176,819 19 January 2000 (19.01.2000) US

(81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

**Published:**

— without international search report and to be republished upon receipt of that report

*For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

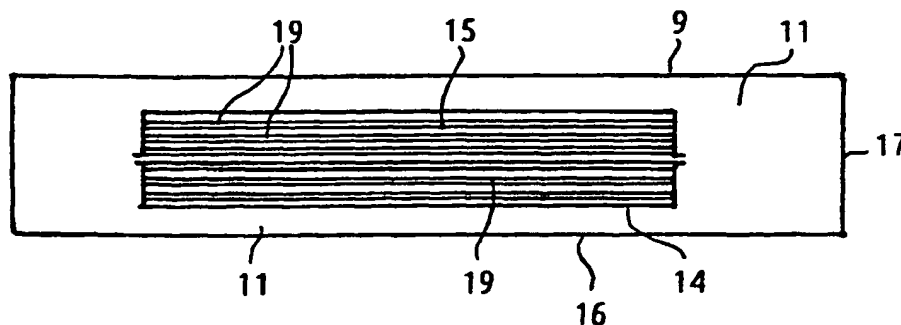
(71) Applicant (*for all designated States except US*): MANHATTAN SCIENTIFICS, INC. [US/US]; Olympic Tower, Suite 36F, 641 Fifth Ave., New York, NY 10022 (US).

(72) Inventor; and

(75) Inventor/Applicant (*for US only*): KOSCHANY, Petra [DE/DE]; Hauptstrasse 6, 94124 Büchlberg (DE).

(74) Agents: RIEDERER VON PAAR, Anton et al.; Freyung 615, 84028 Landshut (DE).

(54) Title: FUEL CELL STACK WITH COOLING FINS AND USE OF EXPANDED GRAPHITE IN FUEL CELLS



(57) Abstract: For an improved cooling a fuel cell stack (1) with cooling fins extending around the periphery of the stack, the section of the stack has a somewhat longitudinal shape. In detail, the stack comprises: a plurality of fuel cells electrically connected in series and having equivalent active section areas and circumferences, each fuel cell comprising a laminate of layers, these comprising an electrolyte membrane and catalyst, electrode and gas diffusion layers the functions of which may be combined in any combination in multifunction layers; end and separator plates delimiting each cell; and cooling layers (9) the cooling function of which also may be combined with other layers in respective multifunction layers, the cooling layers each projecting beyond the circumferential outer periphery (14) of the laminate of the other layers thereby comprising an inner active section area (15) and a peripheral cooling fin area (11). For extending the circumference of the active area (15) and thereby the section of the cooling layer and the area of the fins, the ratio (14:15) of circumference (14) and active area (15) of the stack (1), defined by the geometrical shape of the active area (15), extends the corresponding ratio of a square active area. Also described is the use of expanded graphite for one or several of the following components of a fuel cell: bipolar plate, electrode, and gas diffusion layer.

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**FUEL CELL STACK WITH COOLING FINS  
AND USE OF EXPANDED GRAPHITE IN FUEL CELLS**

**BACKGROUND OF THE INVENTION**

The invention relates to a fuel cell stack comprising: a plurality of fuel cells electrically connected in series and having equivalent active section areas and circumferences, each fuel cell comprising a laminate of layers, these comprising an electrolyte membrane and catalyst, electrode and gas diffusion layers which functions may be combined in any combination in multifunction layers; end and separator plates delimiting each cell; and cooling layers the cooling function of which also may be combined with other layers in respective multifunction layers, the cooling layers each projecting beyond the circumferential outer periphery of the laminate of the other layers thereby an inner active area and a peripheral cooling fin area. The invention further relates to the use of expanded graphite for certain parts of fuel cells.

The need for a cooling system in a hydrogen-air proton exchange membrane (PEM) fuel cell has been a long-standing problem. If the heat rises to a very high temperature level, there exists the danger of drying out the membrane. That results in a loss of ionic conductivity and performance, because of the membrane's need of a high humidity level.

Therefore, fuel cells which have a considerable heat dissipation need a cooling system. For certain applications it is helpful that the cooling system is lightweight and compact, e.g. for a mobile system. In all fuel cell applications the cost factor of an additional peripheral system for the fuel cell system is to be considered.

In U. S. Patent No. 5,595,834, a fuel cell stack is described that has a circular cylindrical shape. For cooling, it uses separator plates that extend

radially outward from the periphery of the stack to serve the additional function of cooling fins. However, the heat transfer to the cooling fins and further to an air stream is restricted which is a considerable limitation particularly for larger cell areas and high temperature gradients and weights. That is because the cooling area of the ring-shaped cooling fins is not sufficient unless using unduely thick layers and broad rings with the consequence of a large overall weight and volume. Rather, the design of the prior art provides relatively large active section areas where waste heat is produced and only relatively small cross sectional areas to conduct the heat outside, and relatively restricted cooling fin areas. An additional cooling system will be necessary that increases the peripheral aggregates of the total system.

U. S. Patent No. 5,776,625 and WO 98/11616 describe similar heat removal schemes. The bipolar plates extend in length or width over the active area of each single cell of the fuel cell stack, forming fins at opposing stack edges or at one stack side, respectively. The concomitant increase in stack surface area allows the stack to be cooled e. g. via air that is blown across its surface by a fan.

In the prior art, no features are disclosed that improve the ratio of the heat dissipation ability to the heat generation in the active area while using thin and lightweight and limitedly large heat conducting layers. A need exists for a cooling system for fuel cell stacks that is inexpensive, lightweight, compact, and conducts all the waste heat of the reaction process out of the fuel cell stack.

For cooling elements, but also for other components of fuel cells, suitable materials are to be found.

## SUMMARY OF THE INVENTION

This invention relates to fuel cell stacks, particularly to those composed of air breathing proton exchange membrane fuel cells, more particularly polymer electrolyte membrane fuel cells, that operate with air as a reactant and cooling gas and have cooling layers comprising external cooling fins. The

cooling layers may be part of the single fuel cells and extend parallelly to the flat fuel cell extension. The purpose is to bring out the total heat of the reaction process, at first from the inside of the stack to the cooling fins and further from the cooling fins to ambient air or another cooling fluid.

5       The fuel cell stack of the invention is characterized in that the ratio of the of circumference and active area of the fuel cells, defined by the geometrical shape of the active area, extends the corresponding ratio of a square active area; or, in other words and rather seen from the perspective of the prism shaped stack, that the ratio of the circumference to the active section area of  
10       the stack, defined by the geometrical shape of the active section area, extends the corresponding ratio of a square base prism, i.e. of the transversal section of a straight parallelepiped with a square base, with the same section area. This provides for a relatively longer circumferencial line around each cell and further for a larger fin area in relation to the fin width, with the consequence of a lower  
15       heat conduction resistance and a higher heat convection.

      The geometrical form of the fuel cells, being members of the fuel cell stack or another configuration of one or more cells itself, is preferably rectangular, or at least has a geometric shape that has a relatively high circumference compared to the area. With this configuration, the distance that the total waste  
20       heat transport has to cover is short, and the cross-section depending on the thickness and the extension of the cooling layer, to conduct the heat outside is high enough even if rather thin cooling layers are used. The smaller distance of the rectangular fuel cell is used as a pathway for the heat. The result of this shape is that the cell, especially the cooling layer, has less weight than if  
25       conventional shapes, like a circle or a square, are used. And as mentioned, for an equal extending width, the circumferencial cooling fin has a larger area for dissipating the heat.

      Though not necessary, it will be assumed favourable to use the separator plates, i. e. the bipolar plates separating the single cells in the stack, for the  
30       cooling plates. However, there is some difficulty in the structure of the bipolar plate. On one hand, the bipolar plate must have enough electric conductivity

orthogonal to the plane of the fuel cell. That is one reason why most fuel cells use a graphite or metal plate as a bipolar plate. But in order to use the bipolar plate as a cooling fin, the heat (or nearby equivalently the electrical) conductivity parallel to the plane of the fuel cell must be enlarged to bring out all the waste heat of the reaction processes. Therefore, in one embodiment the present invention uses a foil containing expanded graphite that has extremely anisotropic features concerning heat and electrical conductivity. The heat conductivity may be fifty (50) times larger parallel to the sheet plane than orthogonal to that plane.

The material that is used for the cooling fin may be rather lightweight. That brings the whole system nearer to the small portable market. A preferred material for use in the cooling layer, according to the invention, is expanded graphite which if also used as a gas diffusion material may be mixed with soot. Expanded graphite is also a useful material for the electrodes of the fuel cell, and for the gas diffusion layer or flow field; the invention also relates to the choices of this material for the mentioned components.

The preferred shape of the active area of the cell is more or less rectangular. Preferably the length is about 1.5 times the width. More preferably, the length is 2.5 to 3.5 times the width.

The cooling layer may be an extension of the anode or cathode flow field or of the bipolar plate or may even be an extension of one electrode. Preferably, it covers the whole active area.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a diagrammatic representation of a fuel cell stack with cooling fins;

Figure 2 is a diagrammatic plan view representation of a cooling fin with gas channels;

Figure 3 is a diagrammatic representation of a heat removing system with a cooling fin and a blower.

## DETAILED DESCRIPTION OF EMBODIMENTS OF THE INVENTION

A polymer electrolyte membrane fuel cell stack 1 consists of a series of single fuel cells 2 separated by bipolar plates 3. Each fuel cell 2 comprises a catalysed membrane 5 to which at both sides a combined gas diffusion and electrode layer 7 is laminated. Between one of the layers 7 of each cell 2 and the associated bipolar plate 3, a cooling layer 9 is inserted that conducts all the waste heat out of the fuel cell, and transfers this heat outside of the cell first to a cooling fin 11 which is a more or less broad rim of the layer 9 outside of the proper stack, and from there to ambient air or another cooling fluid, as shown in Figure 1. End plates 13 form layers at the opposite ends of the fuel cell stack 1.

The geometrical form of the fuel cells 2, being members of the fuel cell stack 1 or of an equivalent configuration of one or more cells, is shown to be longitudinally rectangular, but at least has a geometric shape that has a relatively high circumference 14 compared to the cell area which is the active, transversal section area 15 of the stack. Another possible section shape is e.g. an ellipse. With such configuration, the distance that the total waste heat has to pass through is short, and the cross-section within the cooling layer to conduct the heat outside is high enough even if rather thin cooling layers 9 are used. The small distance from the place of heat generation in the active area 15 to the fin 11 in the rectangular fuel cell is used as a pathway for the heat, and the result of this shape is that the stack 1, and especially its cooling layers 9, has/have less weight than if conventional shapes, like a circle or a square, are used. Further, for an equal width of the circumferential fin 11, the area of the fin 11 relatively to the active area 15 is larger.

The geometrical form of the cooling layers 9 is shown in Figure 2 to also have rectangular shape, and has a length 16 and a width 17. The length 16 of the cooling layer 9 extends the length of the active area 15 of the fuel cells 2, as shown in Figures 1 and 3, and the width 17 of the cooling layer 9 extends the width of the active area 15, leading to the circumferential rim-like cooling fin 11 extending around the cell circumference 14. Figure 2 further shows the

cooling layer 9 to have parallel channels 19 serving as ducts for reaction gas which usually is air which could be pumped into the channels by a (non-depicted) air pump perhaps via a filter. A blower 20 shown in Fig. 3 serves for cooling the cooling fins 11 around the stack 1. The blower 20 can be mounted on an extension of the plate 13 or on some mounting legs, not shown. It transports the cooling air through gaps 22 formed between subsequent cooling fins 11.

Figure 1 shows the fuel cells 2 with the cooling and gas distribution layers 9 on the cathode side. The anode or the cathode or both layers may be designed in that way.

The material of the cooling layers 9 may have the following properties. In order to remove the heat parallel to the cell area, a material is used that has high heat conductivity ( $\lambda$ ), and low density ( $\rho$ ) in order to save weight. A number that characterizes that property of the material is the ratio  $\lambda/\rho$ . Several materials and their properties are given in Table I.

**TABLE I**

Pure Element	Heat Conductivity (W/m K)	Density (g/cm <sup>3</sup> )	$\lambda/\rho$ (W cm <sup>3</sup> /m K g)
Cu	401	8.96	44.8
Ag	429	10.5	40.9
Al	237	2.7	87.8
Ti	21.9	4.5	4.9
Be	201	1.85	108.6
Graphite	150	2.0	75
Foil made of expanded graphite (i.e., Sigrflex from SGL Carbon AG)	165 (parallel) 3 (orthogonal)	1.1	150
Mg	156	1.74	89.6

Ca	125	1.55	80.6
Li	84.8	0.534	158.8

Alkali metals, especially Lithium, have a very desirable  $\lambda/\rho$  ratio, but because of their chemical reactivity and poor mechanical properties they are not desirable as construction materials. Beryllium has very good properties too, but it is rather toxic. Al, Mg and, preferred for the purpose, expanded graphite are some of the remaining materials. Al is a commonly available material. It is important that the Al is pure because Al alloys have a much lower thermal conductivity. However, Al is chemically unstable at fuel cell conditions. A conductive coating may be used, as described in Example 3, to overcome that instability.

The material that has the most benefits is a foil that contains expanded graphite. In order to enhance the mechanical stability and the gas tightness (if needed) of this material, it may be impregnated with conventional resins (epoxy, phenol, phenolic, or furan resin). The electric conductivity of the material is, despite of its anisotropy, high enough to conduct the electric current of the cells. Another layer material that fits very well to the requirements of the cooling layers 9 if at the same time serving as the bipolar plates 3 is a composite material which consists of a metal core, e.g. of Al, Cu, Mg, that is plated with a carbon-polymer layer.

With the extended cooling fins described, it is possible to bring out all the waste heat of the reaction processes. Except for the blower 20, no additional cooling aggregates are necessary. In the special case of a very small fuel cell area, even the blower may be left off. This helps to reduce the peripheral aggregates of the whole system and saves costs.

#### EXAMPLE 1:

In the special form described above the cooling layer 9 also has the function of a gas distribution layer (flow field). In the example, a foil with expanded graphite is used (e. g., Sigrflex from SGL Carbon AG), which is of

a material having an extremely high heat conductivity (see Table I) in the parallel direction and wherein it is easily possible to press or manufacture in another way the gas-channels 19, because the material is soft and deformable due to low content or absence of polymeric binders.

5       The pressing process has two results. One is that the foil is compressed for forming the channels. This means that the density of the material in the channels is higher. For this special material the heat conductivity parallel to the plane of the fuel cell increases with the density of the material. The channels 19 within the cooling layer 9 therefore do not have a negative but rather  
10 a slightly positive effect on the heat conductivity.

      The second result is that all channel structures also have parts, i.e. hills, that are higher than the channels. These parts have the function of conducting the electricity orthogonally to the plane of the cells 2 because of the series connection in the fuel cell stack 1. The electric conductivity decreases in the  
15 direction orthogonally to the plane of the cells 2 with the density of the material, but high conductivity is needed at those parts only where the material is not compressed. So the effect on the paths for the electric conductivity is not a negative one due to such manufacturing process of the channels 19. In summary, in consequence of the channel structure 19, the heat conductivity  
20 is increased, while the electrical conductivity is constant.

      The same arrangement however is possible with other materials (besides expanded graphite) that have a good ratio of density and heat conductivity and that meet the requirements of chemical stability in the fuel cell.

#### 25   EXAMPLE 2:

      In this embodiment of the present invention the cooling layers 9 fulfil the requirements of the bipolar plates 3 being gas separators, too, and thus must be gas-tight. This embodiment may be with or without a channel structure as described above.

30       To overcome the problem with conventional bipolar plate materials that are not useful to bring out all the waste heat parallel to the plane of the cell

because of no enhanced heat conductivity in this direction, the same advanced material as in Example 1 is useful. To fulfil the requirement of gas-tightness, the foil with expanded graphite has to be impregnated or extremely compressed. As an impregnation material a polymer could be used, preferred an epoxy, furan or phenol resin.

In this example, the fuel cells 2 in the stack 1 with the bipolar separator plates 3 also serving as the cooling layers 9 have the following geometrical data:

10	Active area of each cell:	39 mm · 120 mm
	Extension of the cooling fin 1 outside of the circumference 14 and of the active area 15:	20 mm
15	Thickness of the expanded graphite foil:	0.6 mm (i. e., Sigrflex from SGL)
20	Density of the foil (achieved by compression)	1.3 g/cm <sup>3</sup>
	Power density:	0.3 W/cm <sup>2</sup>

The stack 1 has thirty-six (36) of these cells 2 and has a power output of about 500 W. If rather deep channels 19 are required, the thickness of the layer 9 may be enhanced in the active area. The thickness and the mass of the cooling layers 9 still are even smaller than that of liquid cooled systems and all the peripheral aggregates with respect to liquid cooling are eliminated. The cooling layers and the bipolar plates being combined to a multipurpose-layer, the structure of a metal core and a coating layer, e.g. a carbon-polymer layer, serves for a good gas-tightness.

#### EXAMPLE 3:

In another embodiment, the cooling layer 9 again has a double function. In addition to working as a cooling fin, the cooling layer 9 also acts as the electrode. The combined electrode and cooling layer may have the pressed gas

channels 19 as described, and preferably the layer is also gas permeable.

As a preferred material a foil made of expanded graphite is used. The permeability may be reached by using expanded graphite that has a low density, which means that during manufacturing of the foil a low compression force was applied. This gas permeability is only necessary within the active area 15 of the fuel cell 2. To prevent gas losses due to the gas permeability parallel to the foil layer, the material in the cooling fin part 11 may be compressed. Another way to reach a gas impermeability at the cooling fin part 11 (or at least at the transition, i.e. the circumference 14, between active area 15 and cooling fin 11) is to impregnate that part with a polymer, preferably an epoxy, furan or phenol resin.

To reach higher gas permeability in the active area of the foil, in the step before the manufacturing of the foil from the carbon powder mixture the expanded graphite powder may be mixed with a high pore volume soot. The mixing rate may be 90 parts of expanded graphite or less and 10 parts of soot or more, preferred is a ratio of 60 : 40.

To improve the contact between the electrode/cooling layer and the membrane 5, a micro-porous layer may be fitted on the side of the electrode adjacent to the membrane. In Figure 1, the place of such micro-porous layer is shown at 27. Such a micro-porous layer may be made of soot and a hydrophobic material, i. e., polytetrafluoroethylene (PTFE). The connection between the electrode/cooling layer and the micro-porous layer may be obtained by any known coating method, preferably by spraying a suspension onto the surface of the electrode/cooling layer.

#### EXAMPLE 4:

In another embodiment, one or more of the blowers 20 are added to the fuel cell stack 1. They fulfill the task of removing the heat from the cooling fins 11, passing through the gaps 22 formed between subsequent cooling fins 11. The geometrical arrangement of the blowers 20 is important.

Due to the rectangular sectional shape of the fuel cell stack 1, the

pathway for the cooling air 25 parallel to the length is rather long. This results in a relatively high pressure drop within the cooling air path, thereby requiring a high power. To overcome this problem, the flow of the cooling air 25 is divided in at least two directions of pathways 28 and 29 for each long side 16 of the rectangular section, these pathway flows cooling different parts of the cooling fins 11. The flow amount and the flow rate are reduced within each gap 22 formed by the subsequent cooling fins. Therefore the pressure drop is reduced quadratically as a function of the number of pathways 28 and 29. Figure 3 shows such embodiment of the heat removing system, using the cooling fins 11 together with the blower 20 which may be arranged at the width sides 17 of the rectangle as well.

In another embodiment of Example 4, the blower 20 is replaced by the air stream generated due to the running of a vehicle.

Also in another embodiment of the present invention, the blower may be dispensed with. In this embodiment, the cooling air flow results only because of convection. This is possible for rather small stacks or for stacks with low power output per unit active area only.

## Claims:

1. A fuel cell stack (1) comprising: a plurality of fuel cells (2) electrically connected in series and having equivalent active section areas and circumferences, each fuel cell comprising a laminate of layers, these comprising an electrolyte membrane (5) and catalyst, electrode and gas diffusion layers (7) the functions of which may be combined in any combination in multifunction layers; end (13) and separator (3) plates delimiting each cell; and cooling layers (9) the cooling function of which also may be combined with other layers in respective multifunction layers, the cooling layers each projecting beyond the circumferential outer periphery (14) of the laminate of the other layers thereby comprising an inner active section area (15) and a peripheral cooling fin area (11), characterized in that the ratio (14:15) of circumference (14) and active area (15) of the stack (1), defined by the geometrical shape of the active area (15), extends the corresponding ratio of a square active area.

2. The fuel cell stack of claim 1, characterized by an essentially rectangular active section area (15) that has a length (16) exceeding the width (17) by a factor of at least 1.5.

3. The fuel cell stack of claim 2, characterized by an essentially rectangular active section area (15) that has a length (16) exceeding the width (17) by a factor of between 2.5 and 3.5.

4. The fuel cell stack of any of claims 1 to 3, characterized in that the cooling layers, in combination, also are the anodic and/or the cathodic electrodes.

5. The fuel cell stack of any of claims 1 to 3, characterized in that the cooling layers, in combination, also are the anodic and/or the cathodic gas diffusion layers.

6. The fuel cell stack of any of claims 1 to 3, characterized in that the cooling layers, in combination, also are the separator plates (3).

5 7. The fuel cell stack of any of claims 1 to 6, characterized in that the cooling layers (9) have a channel structure in being provided with gas channels (19) within the active section area (15).

10 8. The fuel cell stack of any of claims 1 to 7, characterized in that the material of the cooling layers (9) has a larger specific heat conductivity parallel to the plane of each fuel cell (2) than orthogonal to that plane.

9. The fuel cell stack of claim 8, characterized in that the material of the cooling layers (9) is a foil made of expanded graphite.

15 10. The fuel cell stack of claim 8, characterized in that the material of the cooling layers (9) consists of a metal core that is covered by a carbon-polymer layer.

20 11. The fuel cell stack of any of claims 1 to 10, characterized in that it comprises means (20) for generating an enforced airflow traversing over the cooling fin areas (11) of the cooling layers (9).

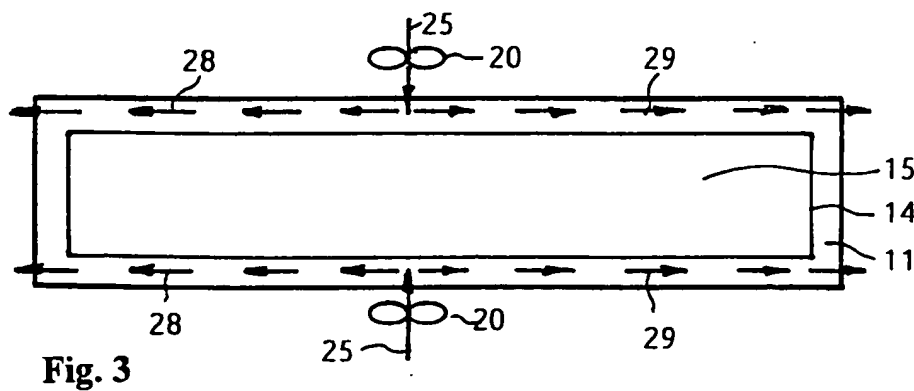
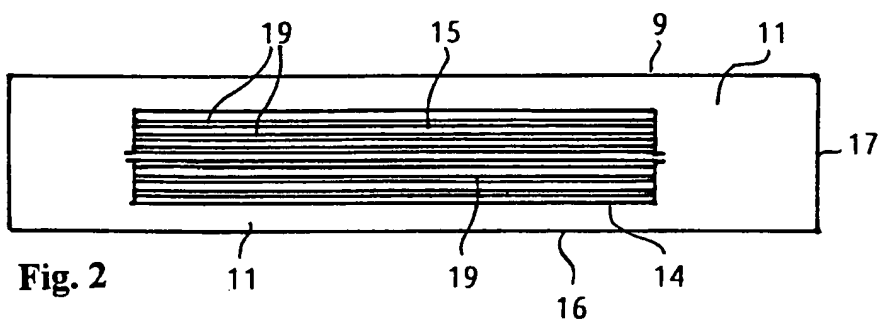
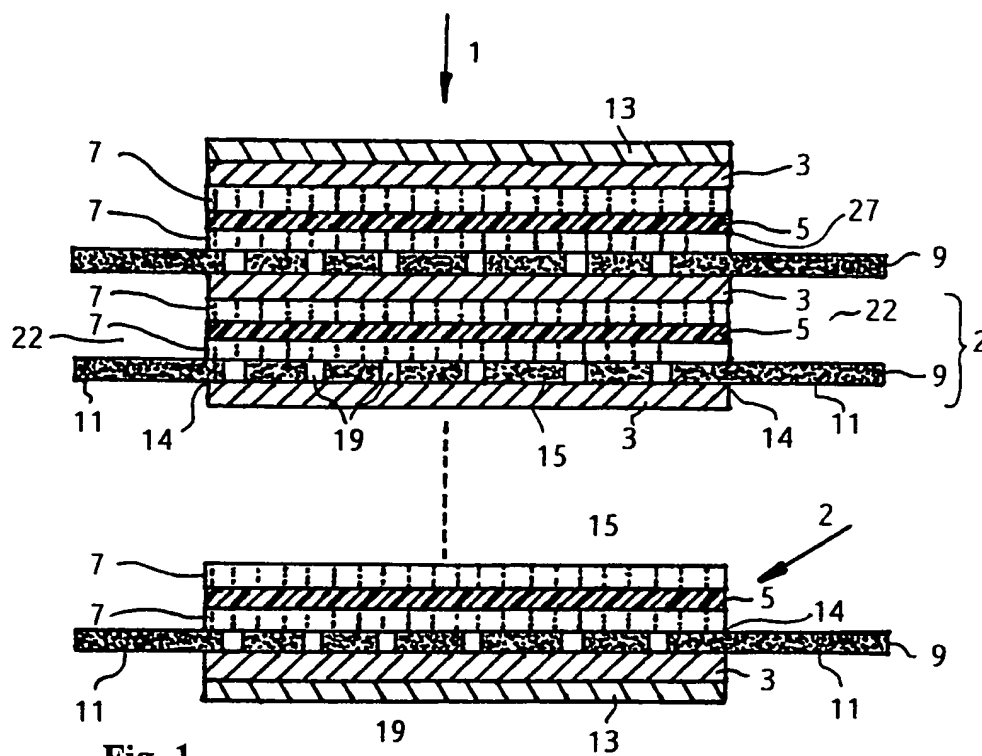
25 12. The fuel cell stack of claim 11 wherein the forced airflow is divided in at least two partial flows (28, 29) each traversing over a part of the cooling fin area (11).

30 13. The fuel cell stack of claim 11 wherein the airflow is supported partly or completely by the dynamic and or static pressure of the ambient air with respect to a moving vehicle.

14. Use of expanded graphite as material for one or several ones of the

following layers of a fuel cell (2) preferably in a fuel cell stack of any of claims 1 to 12: a bipolar plate (3), an electrode (7) and a gas diffusion layer (7).

- 5      15. Use according to claim 14, characterized in that the expanded graphite contains some distributed soot.



(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
26 July 2001 (26.07.2001)

PCT

(10) International Publication Number  
**WO 01/54218 A3**

(51) International Patent Classification<sup>7</sup>: **H01M 8/02**,  
8/24, 8/04, B60L 11/18, H01M 4/96

(21) International Application Number: PCT/EP01/00438

(22) International Filing Date: 16 January 2001 (16.01.2001)

(25) Filing Language: English

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(30) Priority Data:  
60/176,819 19 January 2000 (19.01.2000) US

(71) Applicant (for all designated States except US): **MANHATTAN SCIENTIFICS, INC.** [US/US]: Olympic Tower, Suite 36F, 641 Fifth Ave., New York, NY 10022 (US).

(72) Inventor; and

(75) Inventor/Applicant (for US only): **KOSCHANY, Petra** [DE/DE]: Hauptstrasse 6, 94124 Büchlberg (DE).

(74) Agents: **RIEDERER VON PAAR**, Anton et al.; Freyung 615, 84028 Landshut (DE).

(81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW). Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM). European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR). OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

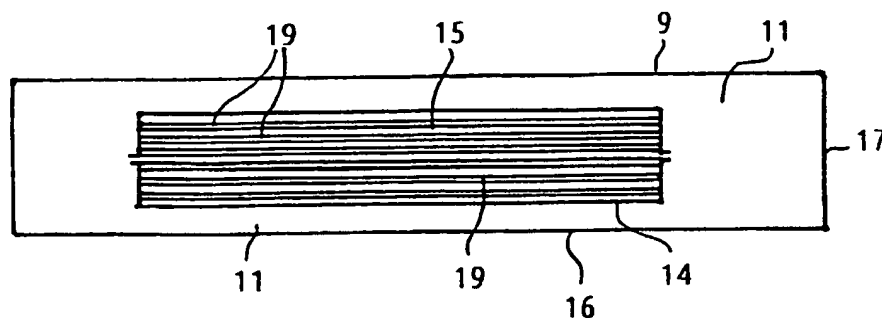
Published:

— with international search report

(88) Date of publication of the international search report:  
21 February 2002

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

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WO 01/54218 A3

# INTERNATIONAL SEARCH REPORT

Int. l. Application No  
PCT/EP 01/00438

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC 7 H01M8/02 H01M8/24 H01M8/04 B60L11/18 H01M4/96		
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) IPC 7 H01M B60L		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
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<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
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<input checked="" type="checkbox"/> Further documents are listed in the continuation of box C. <input checked="" type="checkbox"/> Patent family members are listed in annex.		
* Special categories of cited documents : *A* document defining the general state of the art which is not considered to be of particular relevance *E* earlier document but published on or after the international filing date *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) *O* document referring to an oral disclosure, use, exhibition or other means *P* document published prior to the international filing date but later than the priority date claimed *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. *Z* document member of the same patent family		
Date of the actual completion of the international search  6 November 2001		Date of mailing of the international search report  16/11/2001
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016		Authorized officer  D'hondt, J

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International Application No  
PCT/EP 01/00438

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A	EP 0 343 679 A (INT FUEL CELLS CORP) 29 November 1989 (1989-11-29) column 3, line 26 - line 32 column 5, line 27 - line 34; claim 5; figures 1,2,4 ---	1,6,7,11
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